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DEFOLIATION OF SCOTS PINE AND NORWAY SPRUCE UNDER ALKALINE DUST IMPACT AND ITS RELATIONSHIP WITH RADIAL INCREMENT

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Crown damages of 75–85-year-old Scots pine and Norway spruce within 3 km from the Kunda cement plant, North Estonia, under maximum reported pollution load of 1000–2700 g m⁻² yr⁻¹ in 1985–1999 were assessed in connection with the radial increment of the trees. Strong damages of the trees and their mass dying indicated a long-term pollution load of more than 2 kg m⁻² yr⁻¹, weak and moderate chronic damage of the trees aggravating over years indicated a pollution load of 1–2 kg m⁻² yr⁻¹.

The relations between defoliation and radial increment in the area affected by the cement plant are not linear. A weak defoliation level (needle loss up to 25 %) influenced the radial increment slightly. Correlations were more evident on pine when more than a half of the trees were characterized by moderate and strong defoliation and the percentage of needle loss was thus at least 30–35 %. With a further increase in the defoliation level in the area of a heavier pollution load, the correlation with increment increased both in pines and spruces.

Introduction

Pollutants emitted into the atmosphere by industrial enterprises cause deviations in the metabolism of conifers sensitive to pollutants, such as Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies* (L.) Karst.) [1, 2]. The deviations are expressed also as changes in the state of crowns and production. Air pollution affects trees both directly and through changes in growth environment (soil, precipitation). Pine and spruce react to air pollution depending on the concentration of the pollutants and growth place conditions, also on the genetic characteristics, age and location of the tree within the association. Conifers are particularly sensitive to compounds of sulfur (especially SO₂) and fluor and nitrogen oxides [3, 4]. In the pollutants complex emitted by a cement plant, however, carbonate dust dominates, which does not usually cause acute damage. The influence of cement dust is revealed over a longer period of time being expressed in the physiological-

biochemical processes of the trees, the consequences of which will also become externally visible through morphological changes. It is manifested in a high defoliation level as well as in a decrease in the radial increment of the stem. Low concentrations of cement dust can even stimulate plant growth [5–8].

By forest monitoring, several scientists have acknowledged damages of the crown as an indicator of vitality [9–11]. Disturbance in providing with water and nutrients, changing weather conditions and biotical injuries, which weaken vitality, can turn conifers more sensitive to pollutants [12]. Integratedly, the influence of air pollution is revealed in the increment of conifers. A significant decrease in bioproduction and the death of trees caused by emission of pollutants occur on large areas in the United States of America [13] and Central Europe [14], but also in the surrounding areas of point pollution sources such as power stations, metallurgy enterprises [15, 16], fertilizer manufacturers [17, 18] etc. A significant change in the radial increment of conifers caused by the dust emitted from a cement plant has been established [19, 20].

Some scientists [9, 21, 22] connect the decrease in increment under long-term stress also with changes in the state of the crown, mostly with the increase in defoliation. Investigation of forests near Tallinn in 1978 and 1981 revealed an increase in the defoliation level of conifers and a decrease in the radial increment within about 6 km around the Maardu Chemical Plant [23, 24]. A negative influence of cement dust on the state of crowns and lifetime of needles has also been observed [19, 25].

Applying comparative analysis, the present study explains deviations in the state of the crowns of Scots pine and Norway spruce due to pollutants emitted by a cement plant. We focus on the parameters of defoliation, associating these with the radial increment in areas which are strongly and significantly influenced by the plant.

Study Area and Methods

Growth Conditions in Stands

Climatically, the study area belongs to the mixed-forest subregion of the Atlantic continental region of the temperate zone, which is strongly affected by the vicinity of the Baltic Sea (average yearly temperature is 4.9 °C, minimum 3.2 °C, maximum 7.3 °C; annual amount of precipitation is 550 mm and dominating winds blow from the south and south-west) [26]. Since 1975 the average air temperature and precipitation amount have had rising trends with the average annual air temperature by 0.3 °C and winter temperature by 0.7 °C higher and annual precipitation amount by 41 mm higher over the period 1975–1993 than over a long-term period (1954–1993) [27].

The pollutant emission from the Kunda cement plant (established in 1871, latitude 59°30' N, longitude 26°32' E) varied over the investigation

period depending on the condition of the equipment and the intensity of production, reaching according to the data provided by the plant a maximum in 1991. Dust formed 87–96 % of the total emission, there were small amounts of various exhaust gases (SO_2 , less NO_x , CO and others). As a result of restructuring the production process and the filters launched in 1996, the amount of the pollutants emitted by the plant has considerably decreased. While in 1991 the dust emission in Kunda was 98,600 tons, it was 31,400 tons in 1995 and 14,070 tons in 1996 [28]. According to the data by the cement plant the dust emission is at the minimum level, but as a result of the high pollution level in previous years, the alkalization of the environment is still an essential factor affecting the growth of trees in the influence zone of the cement plant.

The large amounts of cement dust emitted by the plant have, within years, caused notable alkalization of precipitation in the town of Kunda and its vicinity [29]. In 1996, the change in the mean pH of precipitation was slight as compared with the previous years (1996 – 7.62, 1995 – 7.68, 1994 – 7.26), the decrease in several pollution components (Ca, K, Mg, SO_4) was, however, significant [30]. Air pollution has also had a serious impact on soils towards alkalization [31, 32]. The technogenic influence is the strongest in the litter layer of forest soil and it decreases towards deeper layers being in areas with a higher pollution load observable up to the depth of 70 cm. The pH of the humus horizon (maximum 8.5 in 1996) and the concentration of the dominant elements of the cement dust, Ca, K, Mg, S and others, notably exceed the parameters of the control area (30–38 km west from the plant) in spite of the decrease in the pollution load [33, 34].

Study Sites

Studies were carried out in the area of the Kunda cement plant on North-Estonian coastal plain on sample plots stretching 1.5 and 2.5 km east (maximum pollution loads $1800\text{--}2700 \text{ g m}^{-2} \text{ yr}^{-1}$) and 2 and 3 km west ($1000\text{--}1800 \text{ g m}^{-2} \text{ yr}^{-1}$) in 1985–1999. Differences in the crown damages and radial increment of Scots pine and Norway spruce were estimated. The selected stands were similar as to their density, quality class, age, site type and composition of trees (0.7–0.8 density, II quality class, average density or sparse understorey, 75–85-year old *Myrtillus* site type pine or spruce stands), which made comparison of the data possible.

Analysis of Crown Damages and Radial Increment

The state of the crowns was monitored during 15 years (1985–1999). Each autumn the appearance of dominant and co-dominant trees (the type and formation of defoliation, state of the top and mortality of branches, several abiotical and biotical injuries) and the amount and age of needles were estimated. The level of defoliation was established visually and calculated as the percentage of the whole crown compared with the reference (standard) trees

(trees with the maximum crown in the control area). By estimation, a previously-used five-point scale was applied (class I – undamaged tree, defoliation not more than 10 %; class II – weakly damaged tree, defoliation 11–25 %; class III – moderately damaged tree, defoliation 26–60 %; class IV – strongly damaged tree, defoliation more than 60 %; class V – dead tree) [23, 24].

As a result, the damage class of each tree monitored and the number of trees in each class were ascertained. The state of the whole stand was characterized by its damage coefficient, which was calculated by the formula $v = a \times b$, where v is the damage coefficient, a indicates the relative importance of the damage class (%) and b is the damage class. The bigger the product, the greater is the damage and the lower the vitality of the stand. By estimating defoliation, the age of needles (maximal and dominating) of each tree was determined. Annuals of needles where at least 80 % of the needles had remained on a shoot were considered dominating [35].

To study the influence of crown damage on radial increment cores were taken from trees from their northern and southern sides at a height of 1.3 m, and the width of the annual rings was measured. To eliminate the influence of age (trend), the basic data were standardized, i.e. increment indices were calculated on the basis of the actual widths of annual rings [36]. This index shows the ratio of the actual increment and that regarded as standard increment in per cents. To estimate the influence of defoliation on increment, statistical relations between the damage coefficients and indices of radial increment of a stand were found out. The analysis was conducted in the influence zones of the plant. The area up to 2 km west and up to 3 km east of the plant was considered strongly influenced over a longer period of time with the maximal pollution load of 1800–2700 g m⁻² yr⁻¹ (I zone) and the area 2–3 km west and 3–5 km east of the plant was considered significantly influenced, with the pollution load of 1000–1800 g m⁻² yr⁻¹ (II zone) [20].

Results and Discussion

Data on the damages of crowns (defoliation) of conifers in the vicinity of the cement plant are presented in Table 1 and Figures 1 and 2. The relative importance of damaged trees was very high: practically every tree had symptoms of damage. The state of both pines and spruces was the most critical in 1991–1992 (spruces also in 1989), when the damage coefficients were the highest on most sample plots. As to sensitivity towards pollutants, the pine turned out to be more sensitive than the spruce showing a higher level and larger range of damages. In recent years a tendency towards a slow recovery of the state of crowns occurred particularly by the pine, and mostly in the less damaged area, which was still significantly affected by the plant (II zone). Needles of Scots pine remained on the healthy tree up to 3 (4) years, needles of Norway spruce 6–7 (up to 10) years on an average [37].

Table 1. Damage Coefficients* of Pine and Spruce Stands in the Neighbourhood of the Cement Plant in 1985–99

Direction and distance of the research area from the plant, km	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
	Pine stands														
W 2	206	210	212	218	246	256	268	266	259	253	251	252	248	249	245
W 3	225	225	230	231	262	267	278	276	265	259	259	253	254	254	253
E 1.5	240	240	244	251	280	290	308	310	301	300	298	292	290	291	291
E 2.5	244	244	246	253	285	293	305	306	295	296	294	286	286	285	285
	Spruce stands														
W 2	216	216	218	220	231	226	224	225	220	211	210	216	220	218	215
W 3	219	222	222	228	245	237	242	243	238	237	230	231	236	231	230
E 1.5	247	256	255	264	286	277	286	288	284	287	282	288	291	291	293
E 2.5	247	252	254	258	277	270	278	279	273	279	278	283	278	279	281

* 100 = minimum possible and 500 = maximum possible damage.

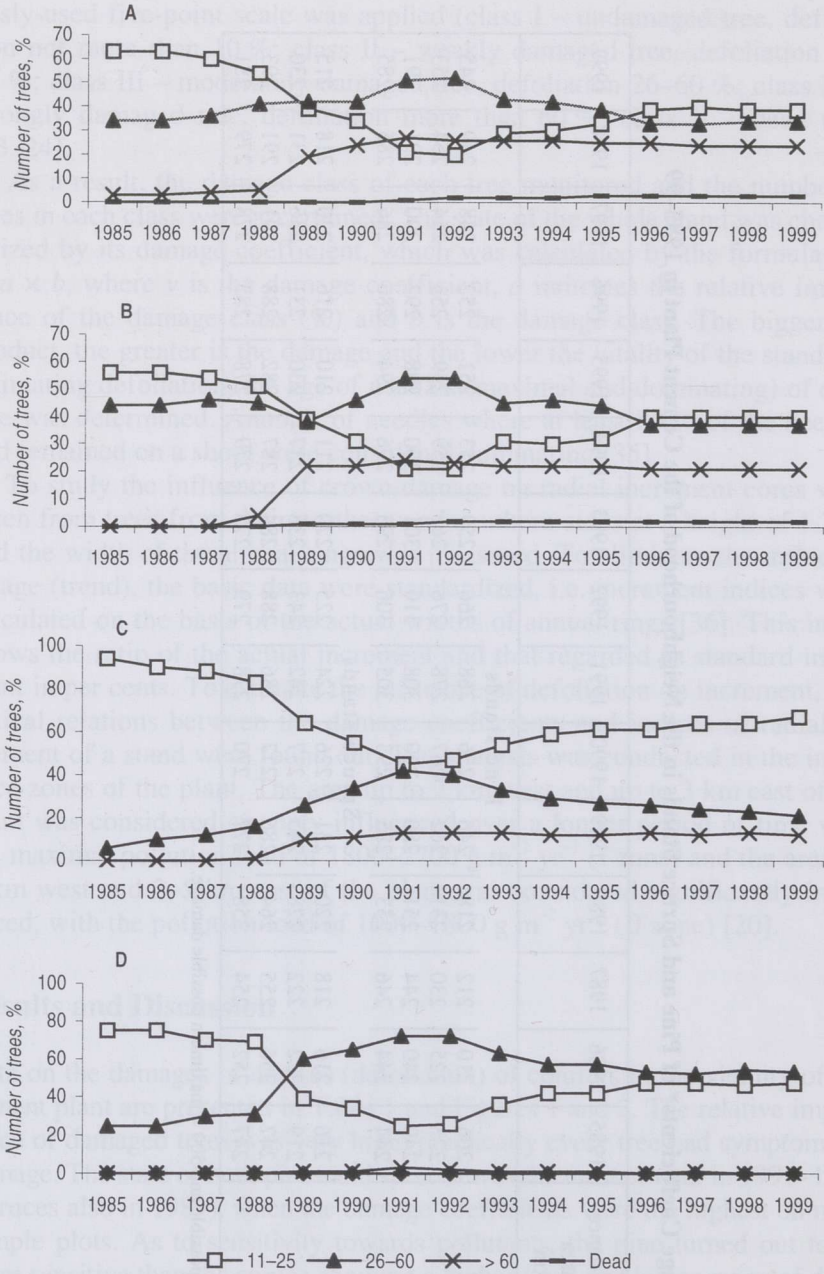


Fig. 1. Number of pine trees by damage classes 1.5 km east (a), 2.5 km east (b), 2 km west (c) and 3 km west (d) from the cement plant in 1985–1999

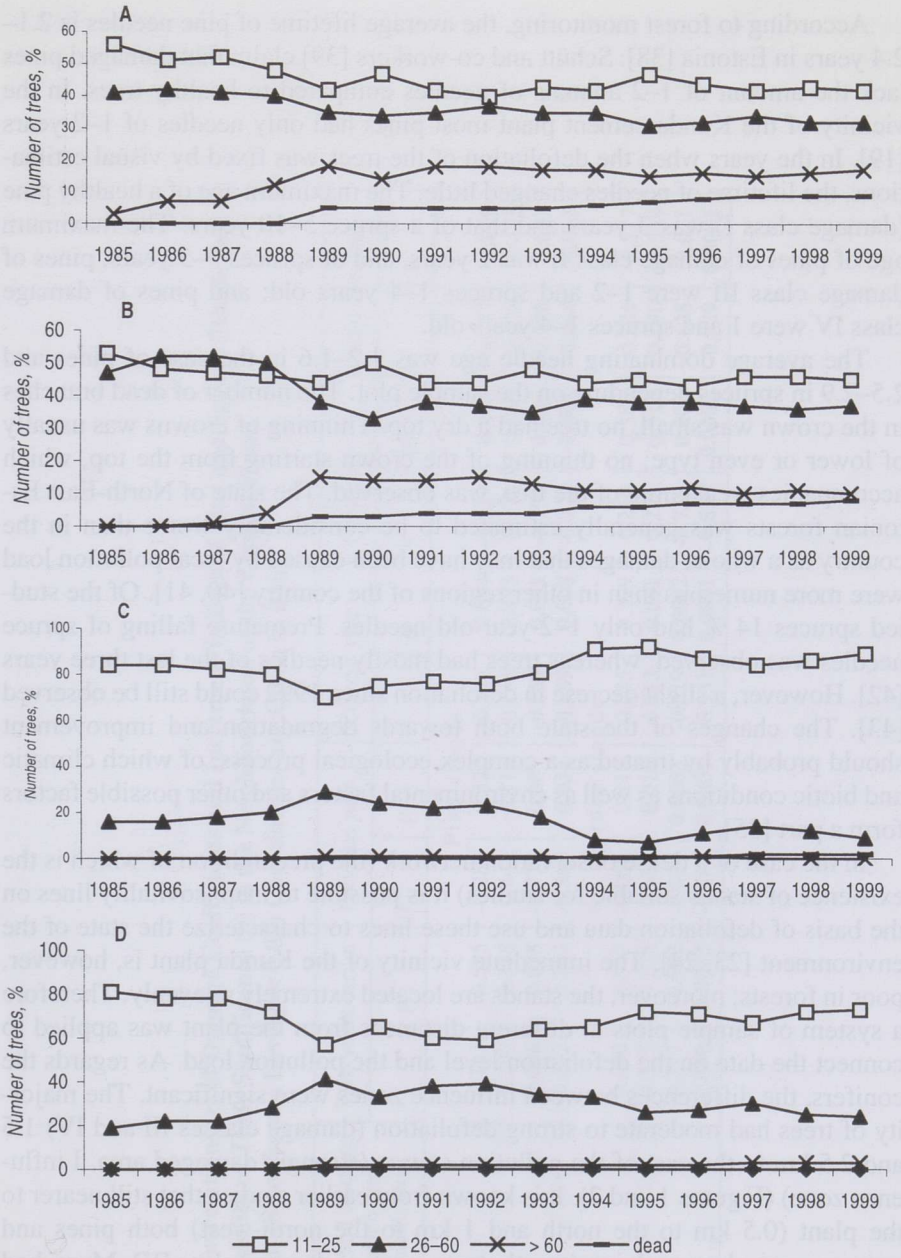


Fig. 2. Number of spruce trees by damage classes 1.5 km east (a), 2.5 km east (b), 2 km west (c) and 3 km west (d) from the cement plant in 1985-1999

According to forest monitoring, the average lifetime of pine needles is 2.1–2.4 years in Estonia [38]. Schütt and co-workers [39] claim that damaged pines lack the amount of 1–2 annuals of needles compared to healthy trees. In the vicinity of the Kunda cement plant most pines had only needles of 1–2 years [19]. In the years when the defoliation of the trees was fixed by visual estimations, the lifetime of needles changed little. The maximum age of a healthy pine (damage class I) was 3 years and that of a spruce 5–10 years. The maximum age of pines of damage class II was 2 years, and of spruces 3–5 years; pines of damage class III were 1–2 and spruces 1–4 years old; and pines of damage class IV were 1 and spruces 1–4 years old.

The average dominating needle age was 1.2–1.6 in the case of pines and 2.5–2.9 in spruces depending on the sample plot. The number of dead branches in the crown was small, no tree had a dry top. Thinning of crowns was usually of lower or even type; no thinning of the crown starting from the top, which accompanies weakening of the tree, was observed. The state of North-East Estonian forests was generally estimated to be considerably worse than in the country as a whole, damages that may have been caused by local pollution load were more numerous than in other regions of the country [40, 41]. Of the studied spruces 14 % had only 1–2-year-old needles. Premature falling of spruce needles was observed, whereas trees had mostly needles of the last three years [42]. However, a slight decrease in defoliation since 1992 could still be observed [43]. The changes of the state both towards degradation and improvement should probably be treated as a complex ecological process, of which climatic and biotic conditions as well as environmental factors and other possible factors form a part [35].

In the case of a dense observation network (the precondition of which is the existence of stands suitable for studies) it is possible to map isovitality lines on the basis of defoliation data and use these lines to characterize the state of the environment [23, 24]. The immediate vicinity of the Kunda plant is, however, poor in forests; moreover, the stands are located extremely unevenly. Therefore a system of sample plots at different distances from the plant was applied to connect the data on the defoliation level and the pollution load. As regards the conifers, the differences between influence zones were significant. The majority of trees had moderate to strong defoliation (damage classes III and IV) 1.5 and 2.5 km to the east of the pollution source (strongly damaged area, I influence zone) (Figures 1 and 2). It is known from earlier studies that still nearer to the plant (0.5 km to the north and 1 km to the north-west) both pines and spruces were close to dying (mostly trees were of damage class IV). Many had already died and had been removed by annual sanitary fells, as proved by the large number of stumps [19]. In the II influence zone (2 and 3 km to the west) the damage coefficients were smaller, and there grew mostly trees of class II. It can be said that strong damages of conifers accompanied by dying out of large numbers of trees under cement dust stress indicates a long-term annual pollution load of more than 2 kg m^{-2} while weak and moderate chronic damages, increasing with time, indicate an annual pollution load of $1\text{--}2 \text{ kg m}^{-2}$.

Table 2 Correlations (r) of Damage Coefficients and Increment Indices of Pine and Spruce Stands and the Probability of Their Significance (p) in the Neighbourhood of the Cement Plant. Significant (Significance Level $p < 0.05$) Correlations are Given in Bold

Direction and distance of the research area from the plant, km	Two years earlier		Previous year		Current year		Next year		After two years	
	r	p	r	p	r	p	r	p	r	p
	Pine stands									
W 2	-0.163	0.595	-0.287	0.320	-0.448	0.094	-0.520	0.054	-0.577	0.039
W 3	0.290	0.338	0.442	0.114	0.196	0.485	0.179	0.540	-0.233	0.445
E 1.5	-0.606	0.028	-0.770	0.001	-0.789	0.001	-0.793	0.001	-0.708	0.007
E 2.5	-0.342	0.253	-0.568	0.034	-0.659	0.008	-0.722	0.004	-0.664	0.013
	Spruce stands									
W 2	0.140	0.647	-0.068	0.818	0.235	0.399	0.250	0.389	0.326	0.277
W 3	0.325	0.278	0.663	0.010	0.513	0.051	0.293	0.310	0.190	0.534
E 1.5	-0.777	0.002	-0.824	0.0003	-0.819	0.0002	-0.736	0.003	-0.628	0.021
E 2.5	-0.840	0.0003	-0.884	0.0001	-0.931	0.0001	-0.838	0.0002	-0.929	0.0001

The high sensitivity of Scots pine and Norway spruce to pollutants emitted by the cement plant has become evident in the changes in radial increment of the trees [44]. On the basis of investigation data a decrease in radial increment, as well as its dependence on the pollution load, can be clearly observed in the zones strongly and significantly affected by the plant (maximum reported load $1000\text{--}2700\text{ g m}^{-2}\text{ yr}^{-1}$). From 1950 up to now the average radial increments of the trees in the monitoring areas up to 2.5 km to the east of the plant usually formed less than 70 % (pines even less than 50 %) of the increments in the control area [27]. In 1985–1999 the difference was even greater (32 % and 41 % of control, respectively). The anthropogenic factor has been dominating in the formation of bioproduction on the study site for a long period of time. The differences in the annual increments of the trees are so small that natural cyclicality of increment is not revealed as a wide amplitude under the given circumstances.

Analysis of the correlation between crown damages and increment provides information on the state of forests and their development. However, the question is with which annual increment the defoliation of the crown is most strongly correlated. It has been assumed that a reduction of radial increment starts already 5–10 years before the appearance of chronic damages [3, 45]. A 5–10 % reduction in radial increment is not accompanied with external injuries of needles, which appear only in the case of 10–20 % increment reduction. When the increment decreases by 20 % or more needle damages are already serious. The decrease in radial increment before damages of needles was also observed by Havas and Huttunen [17] and Kreutzer and others [46]. However, it has also been assumed that increment starts to decrease simultaneously with changes in the crown or 1–2 years after such changes [47]. It is claimed that the relationship between defoliation and increment is not linear and a strong correlation appears only if the level of defoliation is more than 40 % [48]. According to Alexeyev [49] radial increment decreases quickly when the level of defoliation exceeds 60 %. Other authors are, however, of the opinion that even slight defoliation (10–15 %) brings about a significant decrease in increment [50]. It has been concluded that 10 % defoliation decreases increment on an average by 20 % [51], in some cases considerably less – only 11–13 % [52].

Our data on the zone strongly affected by the Kunda cement plant (sample plots 1.5 and 2.5 km to the east from the plant) showed a significant correlation between the level of defoliation of pine and spruce and the radial increment of the current, the next and the next but one year, as well as the preceding year and the year before that (Table 2). The general state of the trees (integratedly shown by increment) determines to a great degree the amount of needles in the crown in the next year and the year after that, this in turn determines the radial increment of the same year and a couple of years to follow. The strongest correlation was observed with the increment of the next year. It is known that the growth of the trees is not influenced solely by the regime of the current vegetation period but always also by the aftereffects of the previous year [36, 53]. The reduced area of assimilating organs and smaller amount of active roots due to

the preceding unfavourable year can decrease increment also during a favourable year. Such aftereffect is especially strong if unfavourable conditions occur several years in succession, or have a synergetic effect.

In the area significantly affected by the plant (sample plots 2 and 3 km to the west) the relationship between defoliation of conifers and radial increment was considerably weaker (Table 2). A sporadical weak correlation occurred between the defoliation level of the pine and the increment of the next year and the year after that, but no such correlation was present by spruces. The ability of the pine and the spruce to compensate for defoliation is different [10]. With regard to photosynthesis, the importance of older needles of spruces is small and their premature falling does not significantly deteriorate the state of the tree.

Summary

In the intensive emission area of the cement plant almost every middle-aged and older conifer bore symptoms of damage. Strong damages of the conifers and their mass dying indicated a long-term pollution load of more than 2 kg m^{-2} per year, weak and moderate cronical damage of the trees, aggravating over years indicated a pollution load of $1\text{--}2 \text{ kg m}^{-2}$ per year. The high sensitivity of Scots pine and Norway spruce to pollutants emitted by the cement plant is further expressed in the changes of the radial increment of the trees. The pine proved to be more sensitive to emission of pollutants than the spruce, being characterized by a higher maximum and larger range of damage. In recent years a tendency of slow recovery of the state of crowns has been observed.

The relationships between defoliation and radial increment in the influence zone of the Kunda cement plant is not linear. A weak defoliation level (needle loss up to 25 %) had a slight correlation with the radial increment. The correlations were stronger on pine when more than a half of the trees were characterized by moderate and strong defoliation and the percentage of needle loss was thus at least 30–35 %. It can be assumed that namely such needle fall accompanied by a decrease in the assimilative area of the trees causes significant changes in the physiological processes. With a further increase in the defoliation level in the zone of heavier pollution, the correlation with increment increased both for pines and spruces.

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Introduction

Separation and fractionation of natural organic matter (NOM) are important topics in environmental and analytical sciences because of NOM role in the carbon cycle and of its influence on heavy metal mobility in water systems. The NOM fraction dissolved organic matter (DOM) constitutes the major part, nearly 90 %, of organic matter in seawater. One part of DOM consists of humic substances (HS). This fraction is present in all natural waters where it constitutes between 10–20 % and more than 80 % of dissolved organic carbon (DOC) for open ocean, and river and lake waters, respectively [1, 2]. HS influence groundwater properties and the process of formation of fossil fuels.